PATENT COOPERATION TREATY

P9881 PCT

PCT

From the INTERNATIONAL BUREAU

To:
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Sett: Vhj 26/6-00

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

Date of mailing (day/month/year)	
15 June 2000 (15.06.00)	
Applicant's or agent's file reference P 9881	**IMPORTANT NOTICE**
International application No. PCT/NO99/00091	**International filing date** (day/month/year) 17 March 1999 (17.03.99)
	Priority date (day/month/year) 04 December 1998 (04.12.98)
Applicant NORSK HYDRO ASA et al	

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:
   AU,CN,JP,KP,KR,US

   In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
   AL,AM,AP,AT,AZ,BA,BB,BG,BR,BY,CA,CH,CU,CZ,DE,DK,EA,EE,EP,ES,FI,GB,GD,GE,GH,GM,HR,
   HU,ID,IL,IN,IS,KE,KG,KZ,LC,LK,LR,LS,LT,LU,LV,MD,MG,MK,MN,MW,MX,NO,NZ,OA,PL,PT,RO,
   RU,SD,SE,SG,SI,SK,SL,TJ,TM,TR,TT,UA,UG,UZ,VN,YU,ZW
   The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on
   15 June 2000 (15.06.00) under No. WO 00/33942

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, **a demand for international preliminary examination** must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the **national phase**, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

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PATENT COOPERATION TREATY

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INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference P 9881	FOR FURTHER ACTION	see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.
International application No. PCT/NO 99/00091	International filing date *(day/month/year)* 17 March 1999	(Earliest) Priority Date *(day/month/year)* 4 December 1998
Applicant NORSK HYDRO ASA et al		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

[X] It is also accompanied by a copy of each prior art document cited in this report.

1. [ ] **Certain claims were found unsearchable** (See Box I).

2. [ ] **Unity of invention is lacking** (See Box II).

3. [ ] The international application contains disclosure of a **nucleotide and/or amino acid sequence listing** and the international search was carried out on the basis of the sequence listing
   - [ ] filed with the international application.
   - [ ] furnished by the applicant separately from the international application,
     - [ ] but not accompanied by a statement to the effect that it did not include matter going beyond the disclosure in the international application as filed.
   - [ ] transcribed by this Authority.

4. With regard to the **title**,
   - [X] the text is approved as submitted by the applicant.
   - [ ] the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,
   - [X] the text is approved as submitted by the applicant.
   - [ ] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is:
   Figure No. 1
   - [ ] as suggested by the applicant.
   - [X] because the applicant failed to suggest a figure.
   - [ ] because this figure better characterizes the invention.
   - [ ] None of the figures.

Method for recovering CO_2

The present invention relates to a method for recovering substantially all CO_2 generated in a combustion process which includes an application of a mixed conducting membrane. Furthermore, the present invention relates to use of this method.

Due to the environmental aspects of CO_2, the possibilities for reducing the emissions of this compound to the atmosphere from combustion processes has been widely discussed.

Conventional combustion processes, used for carbon containing fuels and where the oxygen source is air, having carbon dioxide concentrations of 3-15% in the combustion products, hereinafter called exhaust gas, dependent on the fuel and the applied combustion- and heat recovery process. The reason the concentration is this low is because air is made up of about 78% by volume of nitrogen.

Thus, a reduction in the emission of carbon dioxide to the atmosphare makes it necessary to either separate the carbon dioxide from the exhaust gas, or raise the concentration to levels suitable for use in different chemical processes or for injection and deposition in e.g. a geological formation for long term deposition or for enhanced recovery of oil.

CO_2 can be removed from cooled exhaust gas, normally discharged off at near atmospheric pressure, by means of several separation processes e.g. chemical

active separation processes, physical absorption processes, adsorption by molecular sieves, membrane separation and cryogenic techniques. Chemical absorption for instance by means of alkanole amines is e.g. considered as the most practical and economical method to separate CO_2 from exhaust gas. The separation processes, however, require heavy and voluminous equipment and will consume a substantial amount of heat or power. Applied in a power generation process, this process will reduce the power output with around 10% or more.

An increase of the concentration of CO_2 in the exhaust gas to levels suitable for use in different chemical processes or for injection and deposition e.g. in a geological formation for long term deposition or for enhanced recovery of oil from an oil reservoir is possible by burning fuel in pure oxygen instead of air.

Commercial air separation methods (e.g. cryogenic separation or pressure swing absorption (PSA)) used to produce pure oxygen require 250 to 300 KWh/ton oxygen produced. Supplying oxygen e.g. to a gas turbine by this methods will decrease the net power output of the gas turbine cycle by at least 20%. The cost of producing oxygen in a cryogenic unit will increase the cost of electric power substantially and may constitute as much as 50% of the cost of the electric power.

However, a less energy demanding methode than the separation methods mentioned above is known from European patent application 0658 367- A2 which desribes an application of a mixed conducting membrane which is integrated with a gas turbine system by heating air in a gas turbine combuster and further by selective permeation of oxygen through the membrane. Pure oxygen near atmospheric pressure or below and at high temperature is recovered from the permeate side of the conducting membrane. An oxygen partial pressure difference causes oxygen to be transported through the membrane by reduction of oxygen on the high oxygen partial pressure side (rententate side) and oxidation of oxygen ions to oxygen gas on the low oxygen partial pressure side (the permeate side). In the bulk of the membrane oxygen ions are transported by a diffusion process. Simultaneously the electrons flow from the permeate side back to the feed side of the membrane.

Application of a sweep gas in combination with a mixed conducting membrane to lower the oxygen partial pressure to increase the degree of oxygen removal or oxygen recovery is known from the US patent 5562754. In this patent a method for combined production of oxygen and power is disclosed by heating air in a gas turbine combuster and by selective permeation of oxygen through the membrane. In order to improve the efficiency of gas separation by the membrane, the permeate side of the membrane is swept by e.g. steam supplied for instance from the heat recovery section of the power plant. The sweep gas is heated in a separate high temperature heat exchanger. The application of sweep gas will reduce the partial pressure of oxygen on the permeate side of the membrane and thereby increase the flux of oxygen through the membrane.
However, this require a certain amount of sweepgas and therefore a certain energy amount to generate this sweep gas. This will therefore decrease the net power output of the power generating process.

Application of a sweep gas in combination with a mixed conducting membrane is also known from Norwegian patent application NO-A-972632 (published 07.12.98). This patent describes a power and heat generating process where a fuel is combusted with an oxidant, which is an $O_2/CO_2/H_2O$-containing gaseous mixture, which is supplied from a mixed conducting membrane. The oxygen is picked up from the permeate side of the mixed conducting membrane by means of a sweep gas. The sweep gas is the product or part of the product from at least one combustion process upstream the membrane. In this patent application the sweep gas, or part of the sweep gas, containing a mixture of mainly CO_2 and H_2O, also act as the working fluid in a gas turbine cycle. The amount of sweep gas is related to the amount of working fluid required in the gas turbine cycle i.e. to controll the temperature in the gas turbine combuster. Working fluid is the gas (oxidant and fuel) transported through the gas turbine system. Air fed to the retentate side of this membrane is heated by combusting a fuel in the air stream in a burner.

To obtain a sufficient high flux of oxygen through the membrane a rather high temperature is required (600 -1500 °C). On the air side of the membrane this may

be accomplished by combusting a fuel in the air stream in a burner to increase the temperature of the air fed to the membrane, for instance as disclosed in European patent application 0658 367- A2 or as described in Norwegian patent application NO-A-972632 (published December 7, 1998). The most convenient and least expencive method is to use a carbon containing fuel, e.g. a fossil fuel.
However, by means of this method the heated air stream will contain CO_2 generated in the burner. The CO_2 concentration in the oxygen depleteded air stream discharged from the retentate side of the membrane will be less than about 10% and in most cases less than 3%. If recovery of all generated CO_2 in a combustion process is desirable, due to environmental aspects of CO_2, an oxygen depleted air stream containing low CO_2 -concentrations is not desirable.

Application of a staged mixed conducting membrane process is known from US patent 5,447,555 which describes a method for producing pure oxygen. In this process high purity oxygen is recovered from air by a high-temperature ion transport membrane system comprising two or more stages in which each stage operates at a different feed side to permeate side pressure ratio. Operation of the system in multiple stages at controlled pressure ratios produces oxygen at a lower specific power consumption compared with single-stage operation. Sweep gas is not used in this US patent.

The main object of this invention was to arrive at an energy efficient method to recover substantially all CO_2 generated in a combustion process.

The described object can be fulfilled by application of a method which include an application of a mixed conducting membrane.
Hot steam or a mixture of steam and CO_2 (e.g. recycled exhaust gas) is used as sweep gas to pick up oxygen on the permeate side of a mixed conducting membrane (MCM) in a first stage. The membrane is capable of separating oxygen from a hot air stream fed to the retentate side of the membrane. Sweep gas now containing oxygen is applied as oxidant in a catalytic or non catalytic combustion process where a carbon containing fuel is combusted. Heat generated in the combustion process is used to heat air fed to the retentate side of the membrane.

The hot combustion products, i.e. the exhaust gas, containing CO_2, H_2O and a low concentration of O_2 is used as sweep gas in a second MCM stage and the concentration of oxygen in the sweep gas is increased in the second membrane stage to a sufficiently high level to be used as oxidant in a second combustion stage. Heat generated in the second combustion process is also used to heat air to the MCM-process. Hot combustion products leaving the second combustion stage is used as sweep gas to pick up more oxygen in a third MCM-stage to be used as oxidant in a third combustion stage. The number of required combustion stages and MCM-stages depends on the amount of sweep gas fed to MCM-stage one and on the required pre-heating temperature of air to the retentate side of the MCM-process.

The oxygen produced in the membrane is removed between each stage by combustion with fuel in a combuster. The partially cooled CO_2 containing exhaust gas with a low concentration of oxygen is used as sweep gas in the next MCM stage.

This will reduce the amount of sweep gas necessary for production of a given amount of oxygen and thus reduce the size of equipment necessary for producing sweep gas to the first MCM stage. Application of e.g. 10 stages will reduce the amount of sweep gas with about 95% compared with a single stage process and reduce the energy required to generate sweepgas in the same order of magnitude.

Air fed to the retentate side of each mixed conducting membranes is heated by heat exchanging with hot exhaust gas generated in at least one combuster.

If sweep gas is not generated during the process or used as a working fluid in a gas turbine cycle the sweep gas has to be generated in a separate process.

If sweep gas is generated in a separate process the cost of sweep gas is related to the required amount of sweep gas. The cost of sweep gas generation will be reduced if the amount of sweep gas used in the air heating process is reduced. In a one stage mixed conducting membrane process this reduced amount of sweep gas will, however, reduce the rate of oxygen transport through the membrane. This will further increase the required membrane area and thus the membrane costs. Otherwise the sweep gas pressure has to be reduced. This will, however,

increase the pressure drop of oxygen through the membrane and thus reduce the efficiency of the heat generating process.

In the present invention each stage operates at nearly the same pressure and the staged process will not increase the membrane area requirement. Since oxygen is removed between each stage the driving forces for transport of oxygen through the membrane will increase and reduce the membrane area requirement and the costs.

The problems mentioned above concerning reduced transport of oxygen, increased costs or reduced efficiency, if the amount of sweep gas, which may be steam or a mixture of steam and/or recycled exhaust gas, is reduced, is solved by application of the staged combined mixed conducting membrane and combustion process described in the present invention.
In order to avoid excessive temperatures in the combustion process, comprising use of catalytic or non catalytic combusters, the exhaust gas which is used as sweep gas in a subsequent mixed conducting membrane stage, is cooled between the stages by heat exchanging with air to generate hot air. Furthermore the temperature in the combuster stages are controlled by varying the concentration of oxygen in the sweep gas.

In order to obtain a sufficient high flux of oxygen through the mixed conducting membranes a high air temperature is required which is achieved by the heat exchanging method described above according to the present invention. The air stream is heated in several stages in heat exchangers located between the membrane stages or is divided into several streams and each stream is heated in a heat exchanger located between two membrane stages.

Heated air generated by the method according to the present invention can be used to generate pure oxygen in a mixed conducting membrane.

Furthermore, heated air generated by the method according to the present invention can be used to generate synthesis gas consisting of one or more of the

components CO, CO_2, H_2 and N_2 or for generating heat in a mixed conducting membrane reactor where the membrane reactor is capable of reacting a mixture of steam and a carbon containing fuel with oxygen permeated through the said membrane to make synthesis gas and/or heat.

Further, the method according to the present invention is used in a heat and/or power generating process.

Further, the CO_2-containing exhaust gas generated by the method according to the present invention is used for enhanced oil and natural gas recovery or for injection in a gelogical formation or is used in a chemical process to make carbon containing products. Oxygen eventually left in the CO_2 containing combustion gas exit in the last combustion stage can be removed in a catalytic oxidation reactor or in a combined mixed conducting membrane and partial oxidation reactor as described in patent application NO-A-972631(published 07.12.98).

In a further embodiment of the invention the CO_2 containing exhaust gas and/or the oxygen depleted air stream is depressurized in a turbine to generate power. The invention will be further explained and envisaged in the examples and corresponding figures.

Figure 1 shows a power and heat generating process according to the invention comprising a staged MCM process with inter stage combustion and heat recovery. Air is heated in several stages.

Figure 2 shows a power and heat generating process according to the invention comprising a staged MCM process with inter stage combustion and heat recovery. The air stream from the compressor is divided into several streams and each air stream is heated in a heat exchanger located between two MCM stages.

Figure 1 shows a combined power and heat generating process comprising application of a staged Mixed Conducting Membrane (MCM) and combusters where a sweep gas 1 is fed to the permeate side 2 of a first stage MCM 3 (stage

1) to pick up oxygen transported through said membrane 3. The mixture of sweep gas and oxygen is fed to a catalytic or non-catalytic burner 5 where a fuel 6 is combusted. Hot exhaust gas 7 is fed to a heat exchanger 8 where compressed air 33 is heated. Partially cooled exhaust gas 9 is used as sweep gas on the permeate side 10 of a second MCM 11 (stage 2). Oxygen containing gas 12 is mixed with a fuel 13 in a catalytic or non-catalytic burner 14 to generate a hot exhaust gas 15. Hot exhaust gas 15 with increased amount of CO_2 and H_2O is fed to heat exchanger 16 to heat compressed air 34. Partially cooled exhaust gas 17 is used as sweep gas in a next MCM stage (not shown). The number of required MCM-stages depends on the amount of sweep gas fed to the first MCM-stage. A hot exhaust gas with increased amount of CO_2 and H_2O is used as sweep gas on the permeate side of the last MCM stage 19. Oxygen containing sweep gas 20 is mixed with a fuel 21 in a catalytic or non-catalytic burner 22 to generate a hot exhaust gas 23. This CO_2containing exhaust gas is heat exchanged with compressed air 35 and the CO_2-containing exhaust gas 24 is further depressurized in turbine generator 25 to generate power. Depressurized exhaust gas 26 is fed to heat recovery system 27 to produce steam and condensate water 30. High concentrated CO_2 is recovered and fed to a CO_2 injection system 29.

Air 31 at ambient conditions is fed to compressor 32. Compressed air 33 is further heated in heat exchanger 8 and 16 and further in several stages (not shown) including the last heat exchanger 36. Heated air 37 is fed to the retentate side of MCM 19 and further through several MCM stages including MCM 11 and MCM 3. Partly oxygen depleted air 46 is depressurised in turbine generator 47 to generate power or is fed to a mixed conducting membrane capable of producing pure oxygen or synthesis gas. Depressurised oxygen depleted air 48 is fed to heat recovery system 49 and the cooled gas 50 is discharged off.

Figure 2 shows a combined power and heat generating process comprising application of a staged Mixed Conducting Membrane (MCM) process where a sweep gas 1 is fed to the permeate side 2 of a first stage MCM 3 to pick up oxygen transported through said membrane 3. The mixture of sweep gas and oxygen is fed to a catalytic or non-catalytic burner 5 where a fuel 6 is combusted.

Hot exhaust gas 7 is fed to heat exchanger 8 where compressed air is heated. Partially cooled exhaust gas 9 is used as sweep gas on the permeate side 10 of a second MCM 11 (stage 2). Oxygen containing gas 12 is mixed with a fuel 13 in a catalytic or non-catalytic burner 14 to generate a hot exhaust gas 15. Hot exhaust gas 15 with increased amount of CO_2 and H_2O is fed to heat exchanger 16 to heat compressed air. Partially cooled exhaust gas 17 is used as sweep gas in a next MCM stage (not shown). The number of required MCM-stages depends on the amount of sweep gas fed to MCM-stage one. A hot exhaust gas with increased amount of CO_2 and H_2O is used as sweep gas on the permeate side of a last MCM stage 19. Oxygen containing sweep gas 20 is mixed with a fuel 21 in a catalytic or non-catalytic burner 22 to generate a hot exhaust gas 23. This -CO_2containing exhaust gas is heat exchanged with compressed air 35 and the gas 24 is further depressurized in turbine generator 25 to generate power. Depressurized exhaust gas 26 is fed to heat recovery system 27 to produce steam and condensate water 30. High concentrated CO_2 is recovered and fed to a CO_2 injection system 29.

Air 31 at ambient conditions is fed to compressor 32. Compressed air 33 is further divided into several air streams equal the number of MCM stages. Compressed air stream 35 is heated in heat exchanger 36 and the hot air stream 37 is fed to the retentate side of MCM 19. Oxygen depleted air 38 is fed to mixer 45. Compressed air stream 39 is heated in heat exchanger 16 and the hot air stream 40 is fed to the retentate side of MCM 11. Oxygen depleted air 41 is fed to mixer 45. Compressed air stream 42 is heated in heat exchanger 8 and the hot air stream 43 is fed to the retentate side of MCM 3. Oxygen depleted air 44 is fed to mixer 45. The remaining air streams from separator 51 is fed to the remaining heat exchanger and MCM-stages (not shown) and the resulting hot oxygen depleted air streams is collected in mixer 45. Oxygen depleted air 46 is depressurised in turbine generator 47 to generate power or is fed to a mixed conducting mebrane capable of producing pure oxygen or synthesis gas. Depressurised oxygen depleted air 48 is fed to heat recovery system 49 and the cooled gas 50 is discharged.

Example 1

This example shows a power generating process as described in figure 1.

A hot sweep gas 1 at about 500 to 1200 °C and at elevated pressure consisting of steam or a mixture of steam and CO_2 or recycled exhaust gas (part of stream 28 or stream 24) is fed to the permeate side 2 of a first Mixed Conducting Membrane (MCM) 3 (stage 1) to pick up oxygen transported through said membrane 3. The mixture of sweep gas and oxygen which may contain at least 5 % oxygen is fed to a catalytic or non-catalytic burner 5 where a fuel 6 (i.e. natural gas or synyhesis gas) is combusted. Hot exhaust gas 7 at about 800 to 1500 °C is fed to heat exchanger 8 where compressed air 33 is heated up to between 500 and 750 °C. Partially cooled exhaust gas 9 at 500 to 1200 °C is used as sweep gas on the permeate side 10 of a second MCM 11 (stage 2). Oxygen containing gas 12 is mixed with a fuel 13 in a catalytic or non-catalytic burner 14 to generate a hot exhaust gas 15. Hot exhaust gas 15 at 1000 to 1500 °C with increased amount of CO_2 and H_2O is fed to heat exchanger 16 to heat compressed air 34. Partially cooled exhaust gas 17 is used as sweep gas in a next MCM stage (not shown).

The number of required MCM-stages depends on the amount of sweep gas fed to MCM-stage one. Application of 10 MCM stages will reduce the required amount of sweep gas with about 95% compared with a one stage air heating process as shown in table 1. The amount of sweep gas inlet stage one is 233 kmol/hr and the amount of sweep gas inlet stage 10 is increased to 631 kmol/hr due to the addition of a carbon containing fuel to the burners (5,15, 22 etc.) between each MCM stage and due to addition of oxygen transported through the mixed conducting membranes. The total amount of oxygen produced in all the MCM stages is about 318 kmol/hr if the concentration of oxygen outlet each burner is 3%. By applying 10% oxygen in sweep gas out of a mixed conducting membrane in a one stage process 70% of the recovered oxygen can be used in a combustion process. The amount of sweep gas required to produce 318 kmol/hr of oxygen will then be 4443 kmol/hr. Application of 10 stages will reduce the required amount to 233 kmol/hr i.e. 95% reduction. Table 1 also shows the inlet air temperature of each heat

exchanger (8,16,36 etc.) and the concentration of CO_2 and H_2O in inlet sweep gas of each mixed conducting membrane stage. The CO_2 in the exhaust gas from the last stage may be recovered by cooling the exhaust gas stream to below 50 °C to condensate water. This will increase the concentration of CO_2 to above 95%.
A hot exhaust gas 18 at 1000 to 1300 °C with increased amount of CO_2 and H_2O is used as sweep gas on the permeate side of the last MCM stage 19. Oxygen containing sweep gas 20 is mixed with fuel 21 in a catalytic or non-catalytic burner 22 to generate hot exhaust gas 23. This CO_2-containing exhaust gas at 1100 to 1500 °C is heat exchanged with compressed air 35 and the gas 24 is further depressurized in turbine generator 25 to near atmospheric pressure to generate power. Depressurized exhaust gas 26 is fed to heat recovery system 27 to produce steam and condensate water 30. High concentrated CO2 is recovered and fed to a CO_2 injection system 29 where CO_2 is compressed and dried.

Air 31 at ambient conditions is fed to compressor 32. Compressed air 33 is further heated in heat exchanger 8 and 16 and further in several stages (not shown) including the last heat exchanger 36 up to between 800 and 1400 °C. Heated air 37 is fed to the retentate side of MCM 19 and further through several MCM stages including MCM 11 and MCM 3. Oxygen depleted air 46 is depressurised in turbine generator 47 to near atmospheric pressure to generate power or is fed to a mixed conducting membrane capable of producing pure oxygen or synthesis gas. Depressurised oxygen depleted air 48 is fed to heat recovery system 49 and the cooled nitrogen-containing gas 50 is discharged off.

Table 1

Stage nr.	Inlet Air temperature, °C	%CO_2 in sweep gas inlet stage	%H_2O in sweep gas inlet stage	O_2 transported through MCM kmol/hr
1	454	0	100	26.1
2	461	3.65	93.34	20.9
3	500	6.92	90.06	23.3
4	544	9.86	87.11	25.9
5	592	12.50	84.46	28.9
6	645	14.87	82.08	32.4
7	704	17.01	79.93	36.1
8	769	18.94	77.99	39.6
9	840	20.65	76.28	44.1
10	919	22.18	74.73	49.1
Exit stage 10	1 006	23.56	73.35	

Example 2

This example shows a power generating process as described in figure 2.

The system for generation of sweep gas is the same as described in excample 1 and figure 1. But compressed air 33 is further divided into several air streams equal the number of MCM stages. Compressed air stream 35 is heated in heat exchanger 36 to between 800 and 1400 °C and the hot air stream 37 is fed to the retentate side of MCM 19. Oxygen depleted air 38 is fed to mixer 45. Compressed air stream 39 is heated in heat exchanger 16 to between 800 and 1400 °C and the hot air stream 40 is fed to the retentate side of MCM 11. Oxygen depleted air 41 is fed to mixer 45. Compressed air stream 42 is heated in heat exchanger 8 to between 800 and 1400 °C and the hot air stream 43 is fed to the retentate side of MCM 3. Oxygen depleted air 44 is fed to mixer 45. The remaining air streams from separator 51 is fed to the remaining heat exchanger and MCM-stages (not shown) and the resulting hot oxygen depleted air streams is collected in mixer 45. Oxygen depleted hot air 46 at 800 to 1400 °C is depressurised in turbine generator 47 to generate power or is fed to a mixed conducting mebrane capable of producing

pure oxygen or synthesis gas. Depressurised oxygen depleted air 48 is fed to heat recovery system 49 and the cooled nitrogen-containing gas 50 is discharged.

An alternative configuration of the process acording to figure 1 and 2, comprises that the CO_2 containing gas stream 24 is mixed with a fuel and fed to a catalytic burner in order to remove oxygen. The amount of fuel is regulated such that the concentration of oxygen is reduced to below 50 to 100 ppm. The CO_2 containing exhaust gas with a low concentration of oxygen is depressurised in turbin generator 25 to generate power and heat is further recovered in 27. The CO_2-containing gas after recompression and drying may be injected for enhanced oil recovery.

An alternative configuration of the process according to figure 1 and 2, comprises that the CO_2 containing gas stream 24 is mixed with a fuel and fed to a combined mixed conducting membrane and partial oxidation reactor as described in patent application NO-A-972631 (published 06.12.98) in order to reduce the concentration of oxygen to below 10 ppm. The CO_2 containing exhaust gas with a low concentration of oxygen is depressurised in turbin generator 25 to generate power and heat is further recovered in 27. The -CO_2 containing gas after recompression and drying may be injected for enhanced gas and oul recovery.

An alternative configuration of the process according to figure 1 and 2, comprises that the CO_2 containing gas stream 23 is depressurised in turbine 25 without heatexchanging with air in heat exchanger 36.

Fuel useful in the processes described in example 1 and 2 comprises natural gas, methanol, synthesis gas comprising hydrogen and carbon monoxide, refinery fuel gas containing mixed hydrocarbons or other combustible mixtures.

By the present invention the inventors has arrived at an efficient method to recover substantially all CO_2 generated in a combustion process.

The required amount of sweep gas may be reduced without increasing the required mixed conducting membrane area or without reducing the partial pressure of recovered oxygen. This will reduce the costs and increase the process efficiency.

The method according to the present invention can be used in several types of heat and power generating processes as Combined Cycle Power Plants, in Cogeneration power and heat plants, in Integrated Gasification and Combined Cycle power plants, in any processes comprising combustion of a fuel or for heating purposes e.g. in chemical plants or for heating air fed to mixed conducting membranes.

CLAIMS

1. A method for recovering substantially all carbon dioxide generated in a combustion process,
characterised in that
the method comprises the following steps:

a) a sweep gas is used to pick up oxygen on the permeate side of a mixed conducting membrane in a first_stage which is capable of separating oxygen from a hot air stream fed to the retentate side of the membrane
b) the sweep gas containing oxygen is applied as oxidant in a combuster in the first stage where a carbon containing fuel is combusted
c) hot combustion products of step b) containing CO_2, H_2O and a low concentration of O_2 is used as sweep gas in a membrane in a second stage downstream the combuster in step b)
d) the concentration of oxygen in the sweep gas of step c) is increased in the membrane in the second stage (step c) to a sufficiently high level to be used as oxidant in the combuster in the second stage
e) and the steps c) - d) are repeated in one or more stages.

2. A method for recovering substantially all carbon dioxide generated in a combustion process according to claim 1,
characterised in that
the combuster is a catalytic or non catalytic combuster.

3. A method for recovering substantially all carbon dioxide generated in a combustion process according to claim 1,
characterised in that

the sweep gas used in step a) is hot steam or a mixture of steam and/or recycled exhaust gas from the last combuster in the sequence.

4. A method for recovering substantially all carbon dioxide generated in a combustion process according to claim 1,
characterised in that
the mixed conducting membrane is made from materials with both ionic and electronic conductivity.

5. A method for recovering substantially all carbon dioxide generated in a combustion process according to claim 1,
characterised in that
the air stream is heated by heat exchanging with hot exhaust gas generated in at least one combuster.

6. A method for recovering substantially all carbon dioxide generated in a combustion process according to claim 1,
characterised in that
the air stream, before being heated, is compressed and divided into several streams and each stream is heated in a heat exchanger located between two membrane stages.

7. Use of the method according to claim 1, for generating heat and power.

8. Use of the exhaust gas recovered by the method according to claim 1, for enhanced oil recovery or for injection in a geological formation.

9. Use of the exhaust gas recovered by the method according to claim 1, in a chemical process to make carbon containing products.

10. Use of heated air generated by the method according to claim 1, forgenerating pure oxygen in a mixed conducting membrane.

11. Use of heated air generated by the method according to claim 1, for generating synthesis gas consisting of one or more of the components CO, CO_2, H_2 and N_2 or for generating heat in a mixed conducting membrane reactor where the membrane reactor is capable of reacting a mixture of steam and a carbon containing fuel with oxygen permeated through the said membrane to make synthesis gas and/or heat.

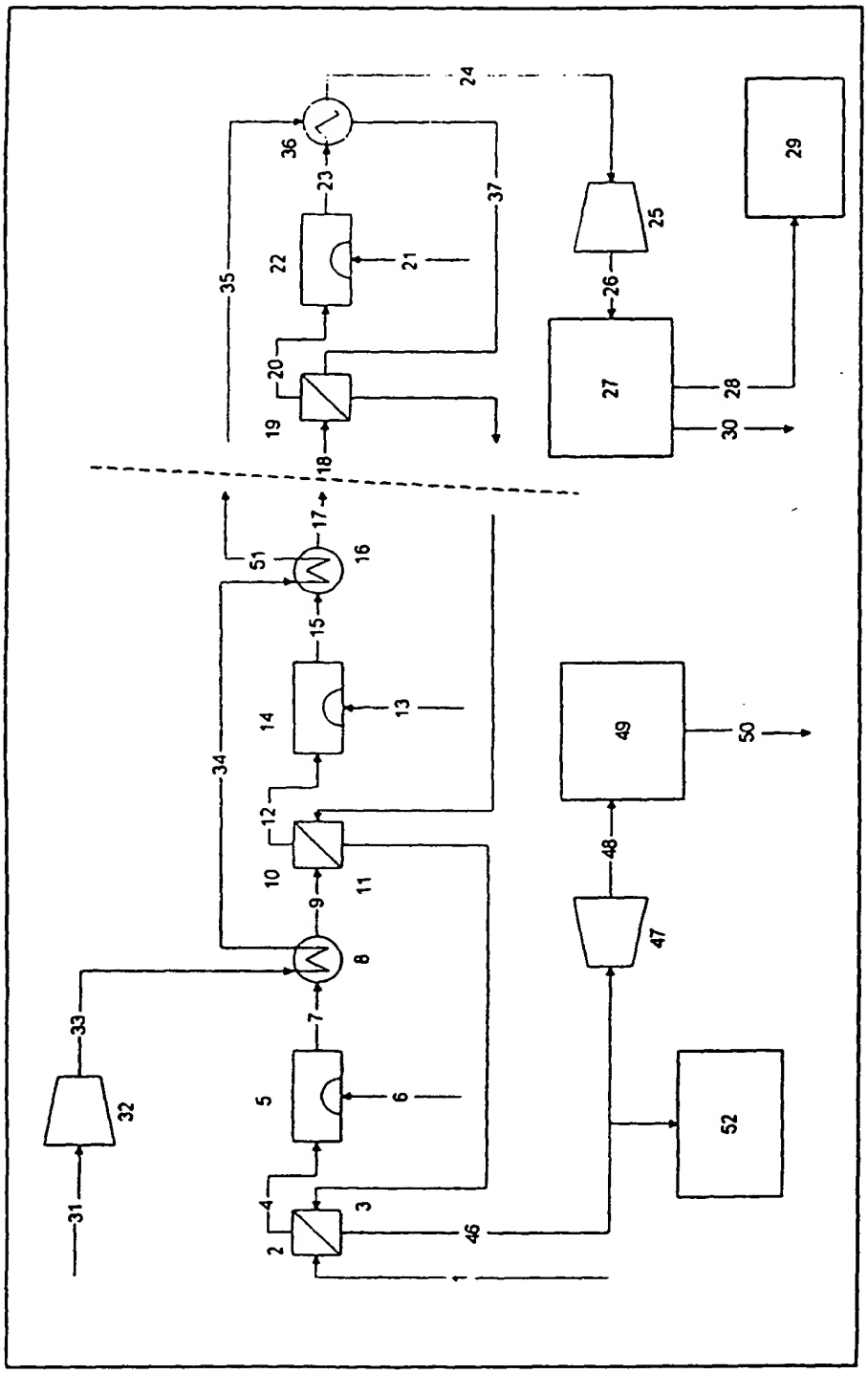
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Figure 1

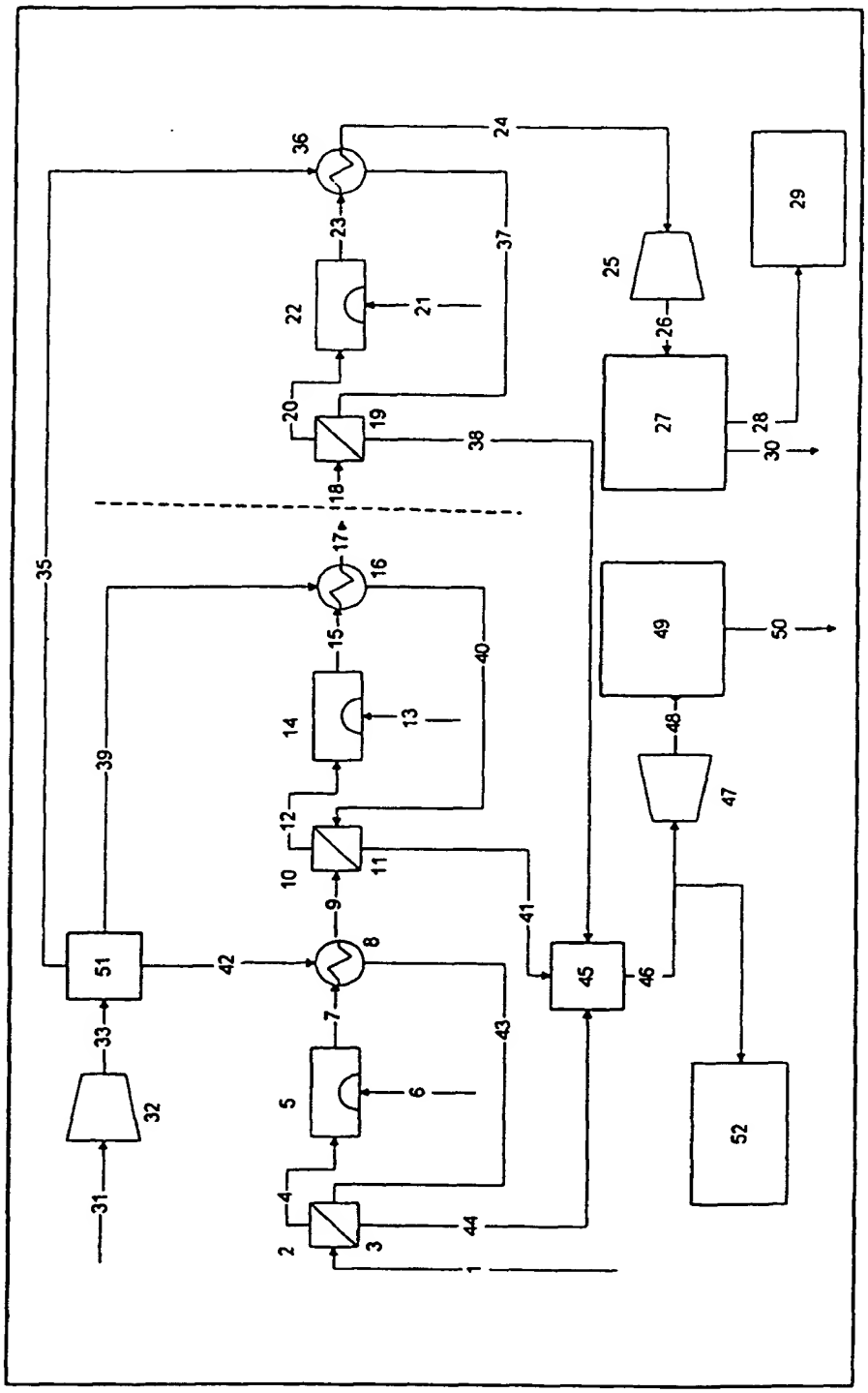
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Figure 2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/NO 99/00091

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: B01D 53/22, C01B 13/02, F23L 7/00
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: B01D, C01B, F23L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI, EPODOC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0778069 A1 (PRAXAIR TECHNOLOGY, INC.), 11 June 1997 (11.06.97), page 4, line 16 - line 58, figure 2, abstract --	1-11
P,A	EP 0882486 A1 (PRAXAIR TECHNOLOGY, INC.), 9 December 1998 (09.12.98), page 4, line 41 - line 50; page 7, line 41 - page 8, line 13, figure 1, claim 1 --	1-11
E	EP 0916386 A1 (PRAXAIR TECHNOLOGY, INC.), 19 May 1999 (19.05.99), figures 6,7, claims 1-10, abstract --	1-11

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" erlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 Sept 1999	08 -10- 1999
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. +46 8 666 02 86	Authorized officer Gunnel Wästerlid/MP Telephone No. +46 8 782 25 00

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/NO 99/00091

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,A	WO 9855208 A1 (NORSK HYDRO ASA), 10 December 1998 (10.12.98), claims 1-10, abstract ---	1-11
A	US 5562754 A (DOOHEE KANG ET AL), 8 October 1996 (08.10.96), claim 1, abstract --- ---	1-11

INTERNATIONAL SEARCH REPORT

Information on patent family members

30/08/99

International application No.
PCT/NO 99/00091

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0778069 A1	11/06/97	AU 704249 B	15/04/99
		AU 7413196 A	12/06/97
		BR 9605848 A	25/08/98
		CA 2192014 A	06/06/97
		CN 1157752 A	27/08/97
		JP 9175802 A	08/07/97
		PL 317307 A	09/06/97
		US 5837125 A	17/11/98
EP 0882486 A1	09/12/98	AU 6992098 A	10/12/98
		CA 2239677 A	05/12/98
		CN 1220181 A	23/06/99
		JP 10339405 A	22/12/98
		US 5888272 A	30/03/99
EP 0916386 A1	19/05/99	AU 9243998 A	10/06/99
		CN 1217586 A	26/05/99
WO 9855208 A1	10/12/98	AU 4139097 A	21/12/98
		NO 972632 A	07/12/98
US 5562754 A	08/10/96	EP 0747109 A	11/12/96

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
P 9881		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/NO99/00091	17/03/1999	04/12/1998
International Patent Classification (IPC) or national classification and IPC B01D53/22		
Applicant NORSK HYDRO ASA ET AL		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 6 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☒ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
22/06/2000	12.03.2001
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Semino, D Telephone No. +49 89 2399 7324

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/NO99/00091

I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*:
Description, pages:

1-14 as originally filed

Claims, No.:

1-11 as originally filed

Drawings, sheets:

1/2-2/2 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-7
	No:	Claims	8-11
Inventive step (IS)	Yes:	Claims	1-7
	No:	Claims	8-11
Industrial applicability (IA)	Yes:	Claims	1-11
	No:	Claims	

2. Citations and explanations
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

Re Item V and VIII

1. It is clear from the description that the following features are essential to the definition of the invention:

i. The sweep gas used in step a) is hot steam or a mixture of steam and/or recycled exhaust gas (dependent claim 3, cf. p. 4, l. 24-26). It is clear from the description that in order for the process to satisfactorily recover the carbon dioxide in a final product, the composition of the sweep stream must satisfy one of the above options; moreover no different options or examples are given in the application.

ii. A carbon containing fuel is fed to a combustor in the first stage **separately** from the sweep gas.

iii. In the second (and following) stage a mixed conducting membrane is used, which is capable of separating oxygen from a hot air stream fed to the retentate side of the membrane, and a combustor is present, to which a carbon containing fuel is separately fed and combusted. Despite of the fact that these features are always implied in the description (cf. e.g. paragraph bridging pages 4 and 5 where it is implied that these features are common to all stages), they are not included in the current wording of claim 1, where they are explicitly mentioned only for the first stage and nothing is said about the further stages being functionally equivalent to the first one (in this respect the wording '**the** combustor' in point d) does not make sense).

Since independent claim 1 does not contain these features it does not meet the requirement following from Article 6 PCT that any independent claim must contain all the technical features essential to the definition of the invention.

2. **Pertinence of the cited prior art**

2.1 Document D1 (EP-A-0778069) discloses (cf. abstract and claim 1) a process for producing a high-purity product from a feed stream containing elemental oxygen by applying the feed stream to a mixed conducting membrane (cf. claim 8) including a feed zone and a permeate zone, and driving a portion of the oxygen contained in the feed stream from the feed zone to the permeate zone via the membrane by applying to the permeate zone a reactive purge stream containing a reactive gas which combines with oxygen to establish a lower pressure of oxygen

in that zone. The reactive gas can be a carbon containing fuel so that a combustion takes place in the permeate zone (cf. p. 3, l. 56 to p. 4, l. 11). In a preferred embodiment, the separator with the membrane may be positioned as a second stage and the feed stream is initially directed to a second feed zone of a second membrane separator, the second separator being positioned as a first stage (cf. p. 3, l. 10-14 and claim 5). The gas exiting the permeate zone of the first separator may be used as a purge stream for the permeate side of the second separator (cf. Figure 5, lines 258 and 260 and p. 7, l. 26-32); a small amount of fuel may be present in such a purge stream (cf. p. 7, l. 17-21).

2.2 Document D2 (US-A-5562754) discloses (cf. claim 1) a process for recovering oxygen from an oxygen-containing gas comprising the steps of compressing and heating a stream of the gas, passing it into a membrane separation zone comprising a mixed conductor membrane (cf. col. 1, l. 36-37), wherein oxygen permeate gas and a hot oxygen-depleted non-permeate gas are produced, introducing water into said hot oxygen-depleted non-permeate gas and passing the resulting gas through an expansion turbine to generate shaft power. Sweeping steam (cf. Figure 2 and claims 13 and 14) can be fed to the permeate side of the membrane separation zone.

3. Conclusions

3.1 The method in claim 1, **modified** so as to include the **essential** features mentioned in paragraph 1, differs from the method disclosed in D1 in that the sweep gas is hot steam or a mixture of steam and/or recycled exhaust gas and in that in each stage fuel is separately fed to a combustor, which is separate from the membrane unit.

3.2 The problem to be solved can be seen as how to provide a method as in D1 in which the recovery of CO_2 is enhanced.

3.3 No hints can be found in the prior art which could prompt the skilled man to modify the method of D1 so as to obtain the method of claim 1, which therefore appears to be novel and inventive (Article 33(2) and (3) PCT) with respect to the available prior art. The same holds for the methods of dependent claims 2-6.

3.4 Claim 7 is incorrectly formulated as an independent use claim. A correct formulation would be: 'A method according to claim 1 characterised in that heat and power are generated'.

3.5 Independent use claims 8-11 concern known uses of known products and are therefore not novel (Article 33(2) PCT). In this respect, the applicants are invited to consider that the exhaust gas and the heated air produced by the method of claim 1 are *per se* not novel, despite of the fact that the process by itself is a novel one. The claims should be deleted.

KOPI til orientering

PCT

REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

For receiving Office use only

PCT/NO 99/00091

International Application No.

17 MARS 1999 (17.03.99)

International Filing Date

PATENTSTYRET
Styret for det industrielle rettsvern
► PCT International application

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference
(if desired) (12 characters maximum) P 9881

Box No. I TITLE OF INVENTION

"Method for recovering CO2"

Box No. II APPLICANT

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)*

NORSK HYDRO ASA
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Norway

☐ This person is also inventor.

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Facsimile No.
+47 22 43 23 08

Teleprinter No.

State *(that is, country)* of nationality: Norway

State *(that is, country)* of residence: Norway

This person is applicant for the purposes of: ☐ all designated States ☒ all designated States except the United States of America ☐ the United States of America only ☐ the States indicated in the Supplemental Box

Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)*

ÅSEN, Knut Ingvar
Tiurveien 22
N-3940 Heistad
Norway

This person is:

☐ applicant only

☒ applicant and inventor

☐ inventor only *(If this check-box is marked, do not fill in below.)*

State *(that is,country)* of nationality: Norway

State *(that is, country)* of residence: Norway

This person is applicant for the purposes of: ☐ all designated States ☐ all designated States except the United States of America ☒ the United States of America only ☐ the States indicated in the Supplemental Box

☒ Further applicants and/or (further) inventors are indicated on a continuation sheet.

Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as: ☒ agent ☐ common representative

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)*

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Norway

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Facsimile No.
+47 22 43 23 08

Teleprinter No.

☐ **Adress for correspondence:** Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

Continuation of Box No. III FURTHER APPLICANTS AND/OR (FURTHER) INVENTORS

If none of the following sub-boxes is used, this sheet should not be included in the request.

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)*

WILHELMSEN, Kjersti
Osebrogata 6
N-3915 Porsgrunn
Norway

This person is:
- ☐ applicant only
- ☒ applicant and inventor
- ☐ inventor only *(If this check-box is marked, do not fill in below.)*

State *(that is, country)* of nationality: Norway

State *(that is, country)* of residence: Norway

This person is applicant for the purposes of: ☐ all designated States ☐ all designated States except the United States of America ☒ the United States of America only ☐ the States indicated in the Supplemental Box

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)*

BRUUN, Tor
Tellusveien 17
N-3942 Skjelsvik
Norway

This person is:
- ☐ applicant only
- ☒ applicant and inventor
- ☐ inventor only *(If this check-box is marked, do not fill in below.)*

State *(that is, country)* of nationality: Norway

State *(that is, country)* of residence: Norway

This person is applicant for the purposes of: ☐ all designated States ☐ all designated States except the United States of America ☒ the United States of America only ☐ the States indicated in the Supplemental Box

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)*

This person is:
- ☐ applicant only
- ☐ applicant and inventor
- ☐ inventor only *(If this check-box is marked, do not fill in below.)*

State *(that is, country)* of nationality:

State *(that is, country)* of residence:

This person is applicant for the purposes of: ☐ all designated States ☐ all designated States except the United States of America ☐ the United States of America only ☐ the States indicated in the Supplemental Box

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)*

This person is:
- ☐ applicant only
- ☐ applicant and inventor
- ☐ inventor only *(If this check-box is marked, do not fill in below.)*

State *(that is, country)* of nationality:

State *(that is, country)* of residence:

This person is applicant for the purposes of: ☐ all designated States ☐ all designated States except the United States of America ☐ the United States of America only ☐ the States indicated in the Supplemental Box

☐ Further applicants and/or (further) inventors are indicated on another continuation sheet.

Box No.V DESIGNATION OF STATES

The following designations are hereby made under Rule 4.9(a) *(mark the applicable check-boxes; at least one must be marked)*:

Regional Patent

- [x] **AP** **ARIPO Patent:** **GH** Ghana, **GM** Gambia, **KE** Kenya, **LS** Lesotho, **MW** Malawi, **SD** Sudan, **SZ** Swaziland, **UG** Uganda, **ZW** Zimbabwe, and any other State which is a Contracting State of the Harare Protocol and of the PCT
- [x] **EA** **Eurasian Patent:** **AM** Armenia, **AZ** Azerbaijan, **BY** Belarus, **KG** Kyrgyzstan, **KZ** Kazakhstan, **MD** Republic of Moldova, **RU** Russian Federation, **TJ** Tajikistan, **TM** Turkmenistan, and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT
- [x] **EP** **European Patent:** **AT** Austria, **BE** Belgium, **CH and LI** Switzerland and Liechtenstein, **CY** Cyprus, **DE** Germany, **DK** Denmark, **ES** Spain, **FI** Finland, **FR** France, **GB** United Kingdom, **GR** Greece, **IE** Ireland, **IT** Italy, **LU** Luxembourg, **MC** Monaco, **NL** Netherlands, **PT** Portugal, **SE** Sweden, and any other State which is a Contracting State of the European Patent Convention and of the PCT
- [x] **OA** **OAPI Patent:** **BF** Burkina Faso, **BJ** Benin, **CF** Central African Republic, **CG** Congo, **CI** C te d Ivoire, **CM** Cameroon, **GA** Gabon, **GN** Guinea, **GW** Guinea-Bissau, **ML** Mali, **MR** Mauritania, **NE** Niger, **SN** Senegal, **TD** Chad, **TG** Togo, and any other State which is a member State of OAPI and a Contracting State of the PCT *(if other kind of protection or treatment desired, specify on dotted line)* .....

National Patent *(if other kind of protection or treatment desired, specify on dotted line)*:

- [x] **AL** Albania .....
- [x] **AM** Armenia .....
- [x] **AT** Austria .....
- [x] **AU** Australia .....
- [x] **AZ** Azerbaijan
- [x] **BA** Bosnia and Herzegovina .....
- [x] **BB** Barbados
- [x] **BG** Bulgaria .....
- [x] **BR** Brazil .....
- [x] **BY** Belarus .....
- [x] **CA** Canada
- [x] **CH and LI** Switzerland and Liechtenstein
- [x] **CN** China .....
- [x] **CU** Cuba .....
- [x] **CZ** Czech Republic .....
- [x] **DE** Germany .....
- [x] **DK** Denmark .....
- [x] **EE** Estonia .....
- [x] **ES** Spain .....
- [x] **FI** Finland .....
- [x] **GB** United Kingdom
- [x] **GD** Grenada
- [x] **GE** Georgia .....
- [x] **GH** Ghana .....
- [x] **GM** Gambia
- [x] **HR** Croatia .....
- [x] **HU** Hungary .....
- [x] **ID** Indonesia
- [x] **IL** Israel .....
- [x] **IN** India .....
- [x] **IS** Iceland
- [x] **JP** Japan .....
- [x] **KE** Kenya .....
- [x] **KG** Kyrgyzstan .....
- [x] **KP** Democratic People s Republic of Korea .....
- [x] **KR** Republic of Korea .....
- [x] **KZ** Kazakhstan .....
- [x] **LC** Saint Lucia
- [x] **LK** Sri Lanka
- [x] **LR** Liberia
- [x] **LS** Lesotho .....
- [x] **LT** Lithuania
- [x] **LU** Luxembourg
- [x] **LV** Latvia
- [x] **MD** Republic of Moldova .....
- [x] **MG** Madagascar .....
- [x] **MK** The former Yugoslav Republic of Macedonia .....
- [x] **MN** Mongolia
- [x] **MW** Malawi .....
- [x] **MX** Mexico .....
- [x] **NO** Norway
- [x] **NZ** New Zealand .....
- [x] **PL** Poland .....
- [x] **PT** Portugal .....
- [x] **RO** Romania
- [x] **RU** Russian Federation .....
- [x] **SD** Sudan
- [x] **SE** Sweden
- [x] **SG** Singapore
- [x] **SI** Slovenia .....
- [x] **SK** Slovakia .....
- [x] **SL** Sierra Leone
- [x] **TJ** Tajikistan .....
- [x] **TM** Turkmenistan .....
- [x] **TR** Turkey .....
- [x] **TT** Trinidad and Tobago .....
- [x] **UA** Ukraine .....
- [x] **UG** Uganda .....
- [x] **US** United States of America .....
- [x] **UZ** Uzbekistan .....
- [x] **VN** Viet Nam .....
- [x] **YU** Yugoslavia .....
- [x] **ZW** Zimbabwe .....

Check-boxes reserved for designating States (for the purposes of a national patent) which have become party to the PCT after issuance of this sheet:

- [ ] .....
- [ ] .....
- [ ] .....

Precautionary Designation Statement: In addition to the designations made above, the applicant also makes under Rule 4.9(b) all other designations which would be permitted under the PCT except any designation(s) indicated in the Supplemental Box as being excluded from the scope of this statement. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. *(Confirmation of a designation consists of the filing of a notice specifying that designation and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.)*

Box No. VI PRIORITY CLAIM ☐ Further priority claims are indicated in the Supplemental Box.

Filing date of earlier application (day/month/year)	Number of earlier application	Where earlier application is: national application: country	regional application:* regional Office	international application: receiving Office
item (1) 04 Dec. 1998; 04.12.98	19985706	Norway		
item (2)				
item (3)				

☒ The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) *(only if the earlier application was filed with the Office which for the purposes of the present international application is the receiving Office)* identified above as item(s): (1)

* *Where the earlier application is an ARIPO application, it is mandatory to indicate in the Supplemental Box at least one country party to the Paris Convention for the Protection of Industrial Property for which that earlier application was filed (Rule 4.10(b)(ii)). See Supplemental Box.*

Box No. VII INTERNATIONAL SEARCHING AUTHORITY

Choice of International Searching Authority (ISA) *(if two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used):*

ISA / SE

Request to use results of earlier search; reference to that search *(if an earlier search has been carried out by or requested from the International Searching Authority):*

Date *(day/month/year)* Number Country *(or regional Office)*

Box No. VIII CHECK LIST; LANGUAGE OF FILING

This international application contains the following **number of sheets:**

request	: 4
description (excluding sequence listing part)	: 14
claims	: 3
abstract	: 1
drawings	: 2
sequence listing part of description	:
Total number of sheets	: 24

This international application is **accompanied by** the item(s) marked below:

1. ☒ fee calculation sheet
2. ☐ separate signed power of attorney
3. ☐ copy of general power of attorney; reference number, if any:
4. ☐ statement explaining lack of signature
5. ☐ priority document(s) identified in Box No. VI as item(s):
6. ☐ translation of international application into (language):
7. ☐ separate indications concerning deposited microorganism or other biological material
8. ☐ nucleotide and/or amino acid sequence listing in computer readable form
9. ☐ other *(specify)*:

Figure of the drawings which should accompany the abstract: 1

Language of filing of the international application: English

Box No. IX SIGNATURE OF APPLICANT OR AGENT

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request).

Venche H. Johnsen

Venche Høines Johnsen

For receiving Office use only

1. Date of actual receipt of the purported international application: 17 MARS 1999 (17.03.99)
2. Drawings: ☒ received: ☐ not received:
3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application:
4. Date of timely receipt of the required corrections under PCT Article 11(2):
5. International Searching Authority (if two or more are competent): ISA / SE
6. ☐ Transmittal of search copy delayed until search fee is paid.

For International Bureau use only

Date of receipt of the record copy by the International Bureau:

INTERNATIONAL SEARCH REPORT

International application No.
PCT/NO 99/00091

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: B01D 53/22, C01B 13/02, F23L 7/00
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: B01D, C01B, F23L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI, EPODOC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0778069 A1 (PRAXAIR TECHNOLOGY, INC.), 11 June 1997 (11.06.97), page 4, line 16 - line 58, figure 2, abstract --	1-11
P,A	EP 0882486 A1 (PRAXAIR TECHNOLOGY, INC.), 9 December 1998 (09.12.98), page 4, line 41 - line 50; page 7, line 41 - page 8, line 13, figure 1, claim 1 --	1-11
E	EP 0916386 A1 (PRAXAIR TECHNOLOGY, INC.), 19 May 1999 (19.05.99), figures 6,7, claims 1-10, abstract --	1-11

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" erlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 Sept 1999	08 -10- 1999
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. + 46 8 666 02 86	Authorized officer Gunnel Wästerlid/MP Telephone No. + 46 8 782 25 00

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/NO 99/00091

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,A	WO 9855208 A1 (NORSK HYDRO ASA), 10 December 1998 (10.12.98), claims 1-10, abstract ---	1-11
A	US 5562754 A (DOOHEE KANG ET AL), 8 October 1996 (08.10.96), claim 1, abstract --- ---	1-11

INTERNATIONAL SEARCH REPORT

Information on patent family members

30/08/99

International application No.
PCT/NO 99/00091

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0778069 A1	11/06/97	AU 704249 B	15/04/99
		AU 7413196 A	12/06/97
		BR 9605848 A	25/08/98
		CA 2192014 A	06/06/97
		CN 1157752 A	27/08/97
		JP 9175802 A	08/07/97
		PL 317307 A	09/06/97
		US 5837125 A	17/11/98
EP 0882486 A1	09/12/98	AU 6992098 A	10/12/98
		CA 2239677 A	05/12/98
		CN 1220181 A	23/06/99
		JP 10339405 A	22/12/98
		US 5888272 A	30/03/99
EP 0916386 A1	19/05/99	AU 9243998 A	10/06/99
		CN 1217586 A	26/05/99
WO 9855208 A1	10/12/98	AU 4139097 A	21/12/98
		NO 972632 A	07/12/98
US 5562754 A	08/10/96	EP 0747109 A	11/12/96

PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

To:

Assistant Commissioner for Patents
United States Patent and Trademark Office
Box PCT
Washington, D.C.20231
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 02 August 2000 (02.08.00)	
International application No. PCT/NO99/00091	**Applicant's or agent's file reference** P 9881
International filing date (day/month/year) 17 March 1999 (17.03.99)	**Priority date** (day/month/year) 04 December 1998 (04.12.98)
Applicant ÅSEN, Knut, Ingvar et al	

1. The designated Office is hereby notified of its election made:

[X] in the demand filed with the International Preliminary Examining Authority on:

22 June 2000 (22.06.00)

[ ] in a notice effecting later election filed with the International Bureau on:

2. The election [X] was

[ ] was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer Claudio Borton Telephone No.: (41-22) 338.83.38

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NO9900091

TENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION CONCERNING SUBMISSION OR TRANSMITTAL OF PRIORITY DOCUMENT

(PCT Administrative Instructions, Section 411)

To:

JOHNSEN, Venche, Høines
Norsk Hydro ASA
N-0240 Oslo
NORVÈGE

Date of mailing (day/month/year) 31 May 1999 (31.05.99)	
Applicant's or agent's file reference P 9881	**IMPORTANT NOTIFICATION**
International application No. PCT/NO99/00091	International filing date (day/month/year) 17 March 1999 (17.03.99)
International publication date (day/month/year) Not yet published	Priority date (day/month/year) 04 December 1998 (04.12.98)
Applicant NORSK HYDRO ASA et al	

1. The applicant is hereby notified of the date of receipt (except where the letters "NR" appear in the right-hand column) by the International Bureau of the priority document(s) relating to the earlier application(s) indicated below. Unless otherwise indicated by an asterisk appearing next to a date of receipt, or by the letters "NR", in the right-hand column, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).

2. This updates and replaces any previously issued notification concerning submission or transmittal of priority documents.

3. An **asterisk(*)** appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b). In such a case, **the attention of the applicant is directed** to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

4. The **letters "NR"** appearing in the right-hand column denote a priority document which was not received by the International Bureau or which the applicant did not request the receiving Office to prepare and transmit to the International Bureau, as provided by Rule 17.1(a) or (b), respectively. In such a case, **the attention of the applicant is directed** to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

Priority date	Priority application No.	Country or regional Office or PCT receiving Office	Date of receipt of priority document
04 Dece 1998 (04.12.98)	19985706	NO	18 May 1999 (18.05.99)

The International Bureau f WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer

Aino Metcalfe

Telephone No. (41-22) 338.83.38